

# Optical Manipulation of Magnetism in Correlated Electron Systems

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Thanks to Koshihara, Okimoto, Iwai



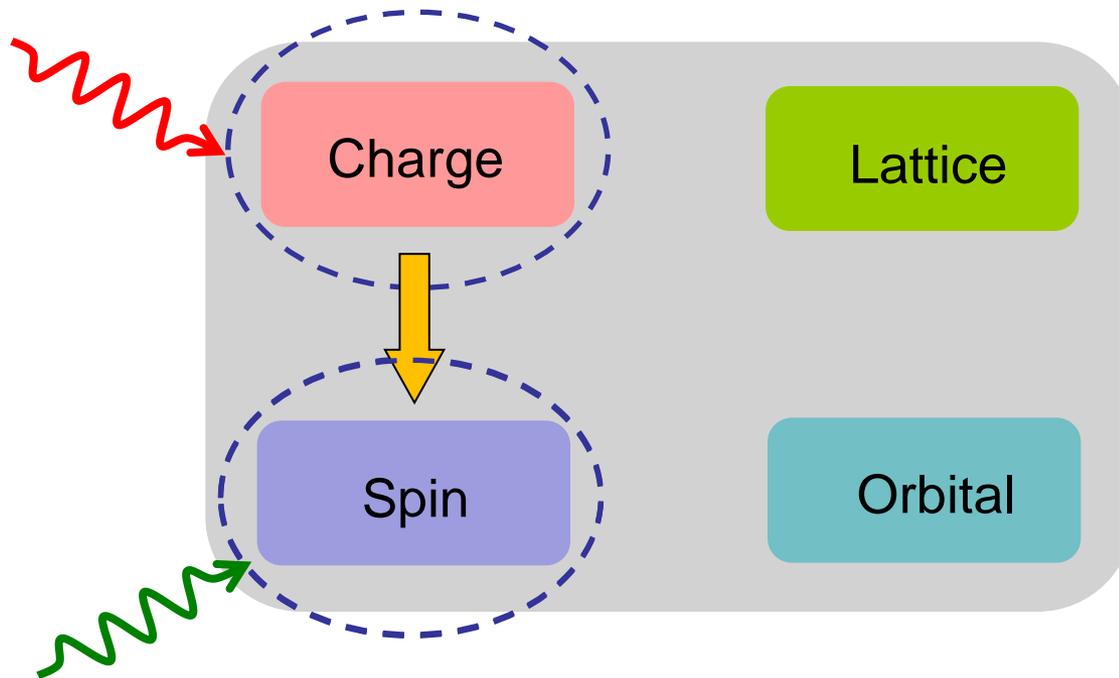
TOHOKU  
UNIVERSITY

IMPACT @ Paris-sud, Orsay  
September 10-14, 2012



# Spin-charge coupled system

Optical manipulation of magnetism  
in spin-charge coupled system

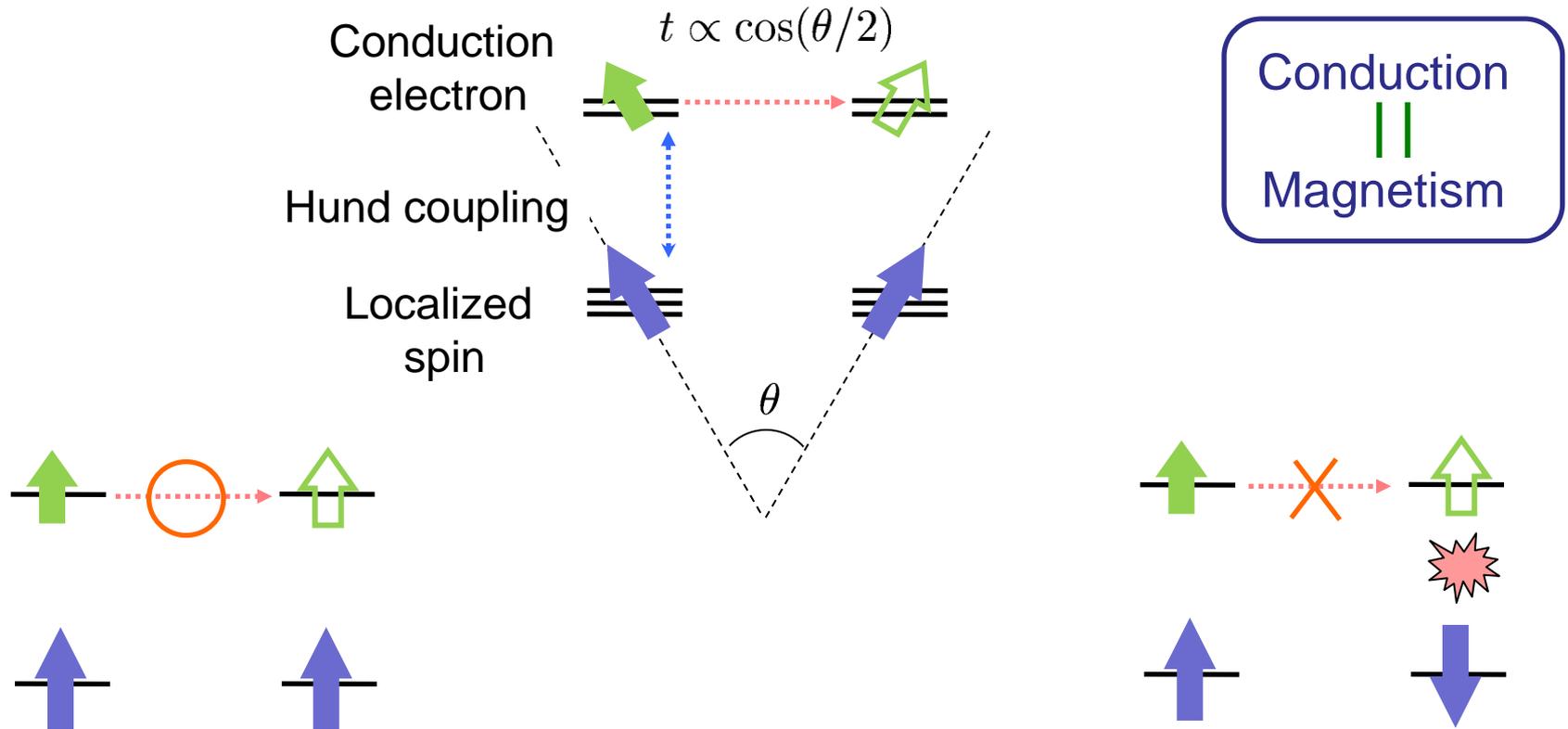


Circular polarized light

Spin-orbit coupling

# Double exchange interaction

Zener ('51), Anderson-Hasegawa ('55), de Gennes ('59)



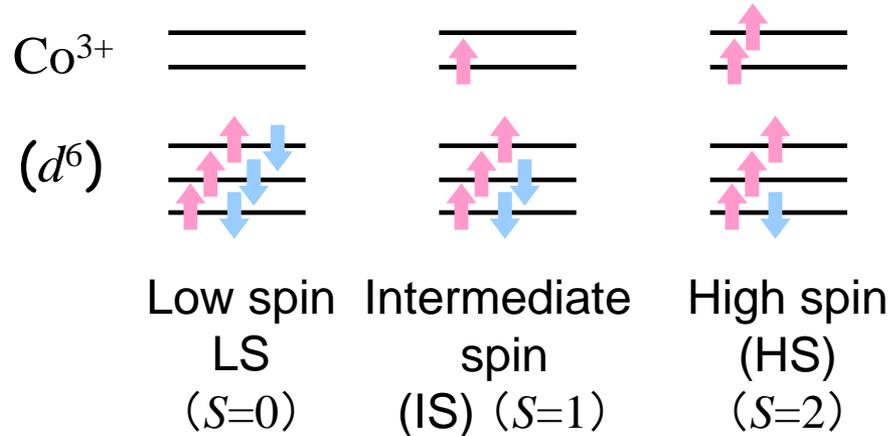
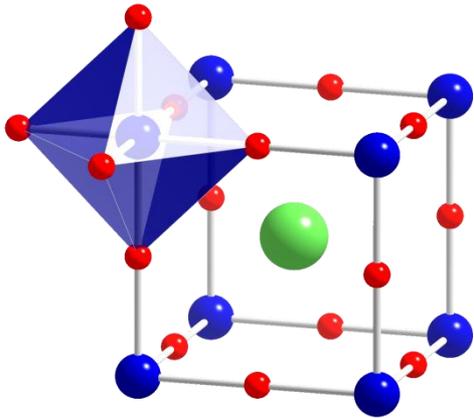
Present talk

Spin-state transition (Cobaltites)  
Inverse Double-exchange phenomena (Manganites)

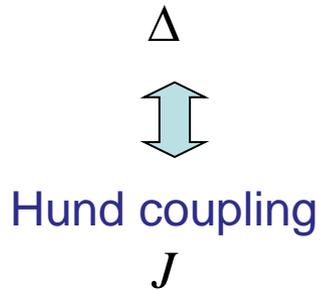
Photo-induced HS bound state  
in spin-transition system  
(Cobaltites)

# Perovskite cobaltites

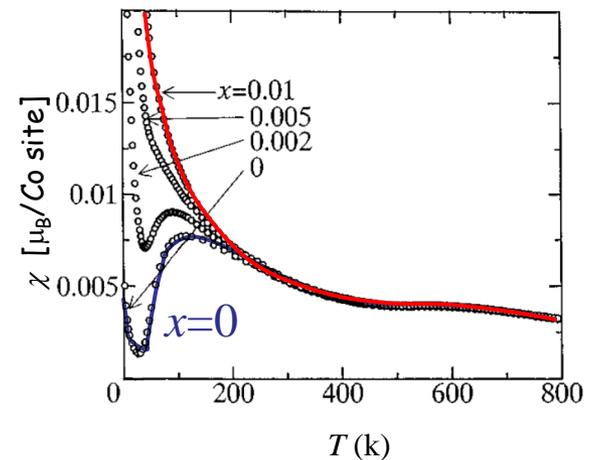
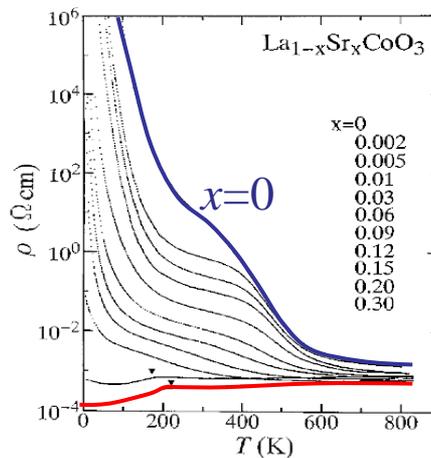
LaCoO<sub>3</sub>



Crystalline field splitting



Spin transition by temperature, and doping

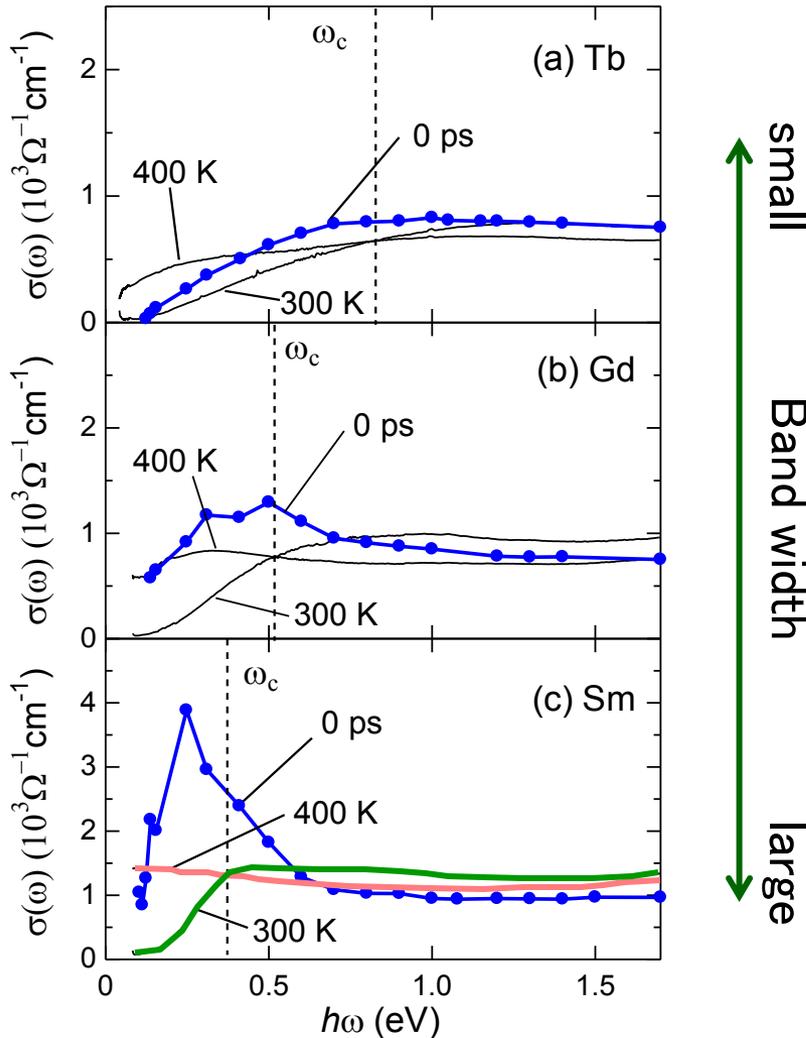


Y. Tokura *et al.* Phys. Rev. B ('98)

# Photo irradiation



Prof. Koshihara's talk



- Non-magnetic insulator



Metallic

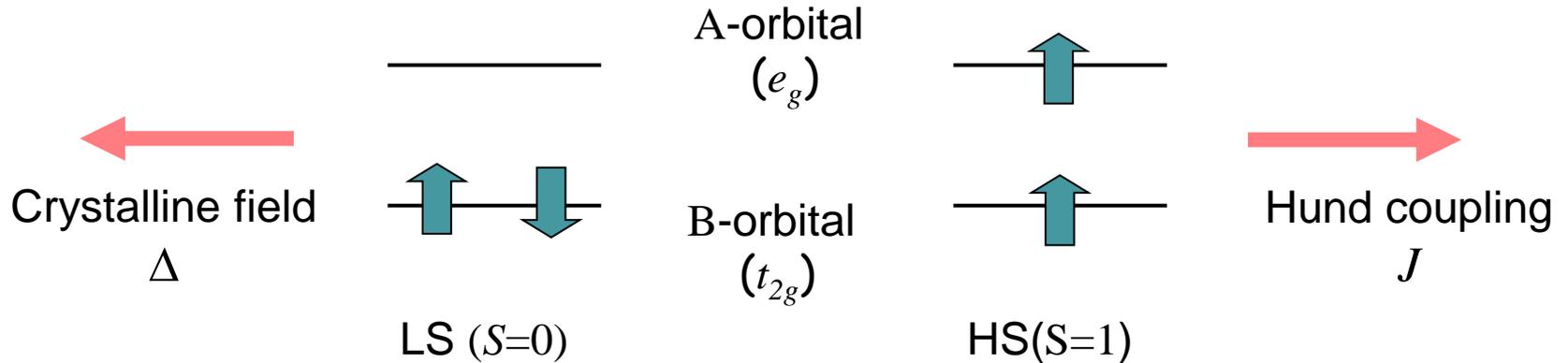
- Photo excitation



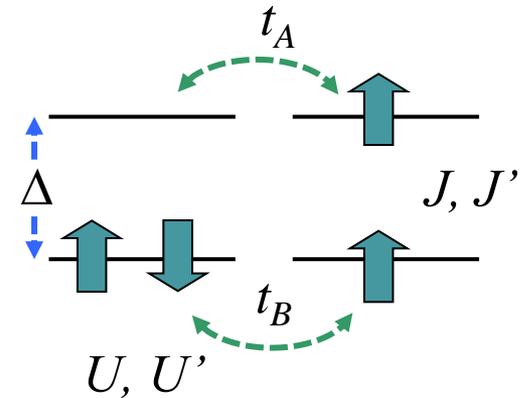
Thermal excitation

- $R$ -ion (band width) dependence

# Theoretical model



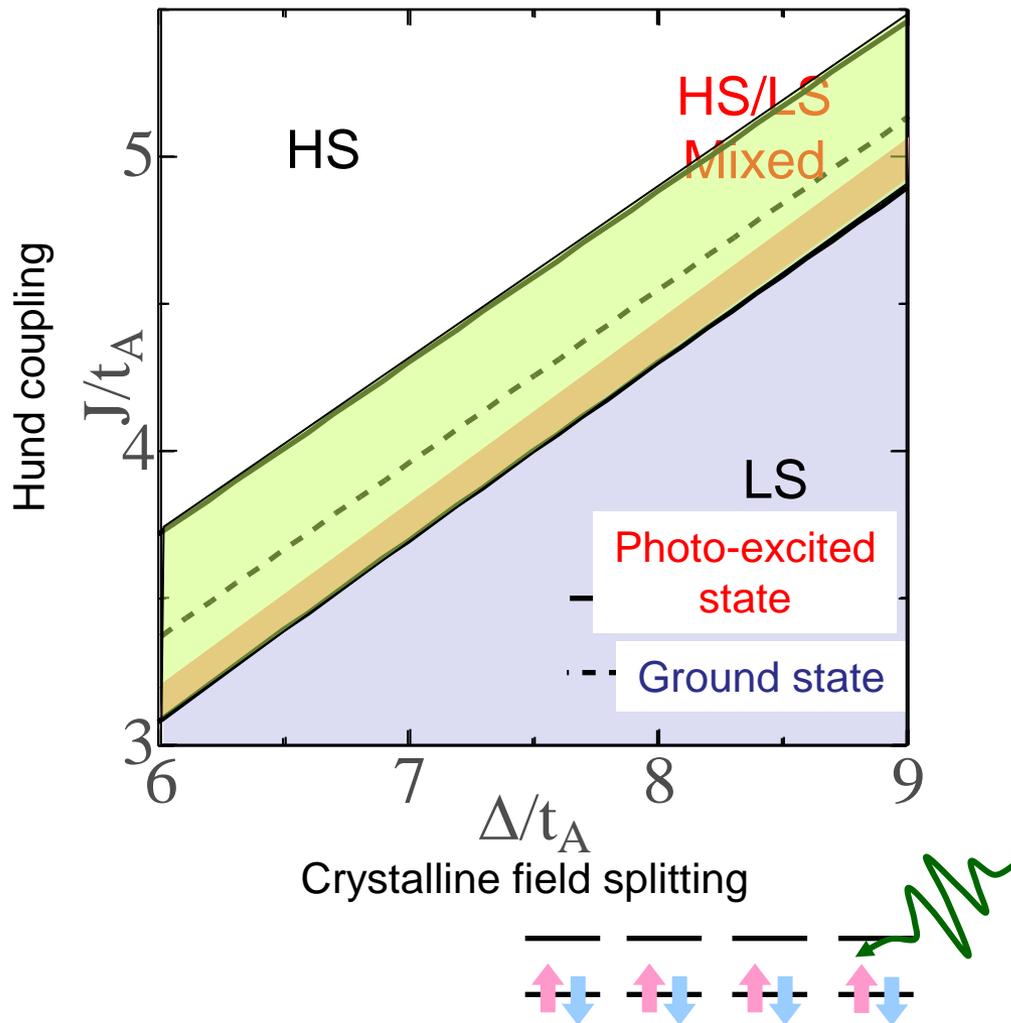
$$\begin{aligned}
 \mathcal{H} = & \Delta \sum_{i\sigma} c_{iA\sigma}^\dagger c_{iA\sigma} - \sum_{\langle ij \rangle \gamma \sigma} t_\gamma (c_{i\gamma\sigma}^\dagger c_{j\gamma\sigma} + H.c.) \\
 & + U \sum_{i\gamma} n_{i\gamma\uparrow} n_{i\gamma\downarrow} + U' \sum_{i\sigma\sigma'} n_{iA\sigma} n_{iB\sigma'} \\
 & - J \sum_{i\sigma\sigma'} c_{iA\sigma}^\dagger c_{iB\sigma} c_{iB\sigma'}^\dagger c_{iA\sigma'} - J' \sum_{i\gamma} c_{i\gamma\uparrow}^\dagger c_{i\bar{\gamma}\uparrow} c_{i\gamma\downarrow}^\dagger c_{i\bar{\gamma}\downarrow},
 \end{aligned}$$



Ave. electron # /site = 2

Exact diagonalization & t-dependent HF

# Photo-induced spin state change



■ LS band insulator

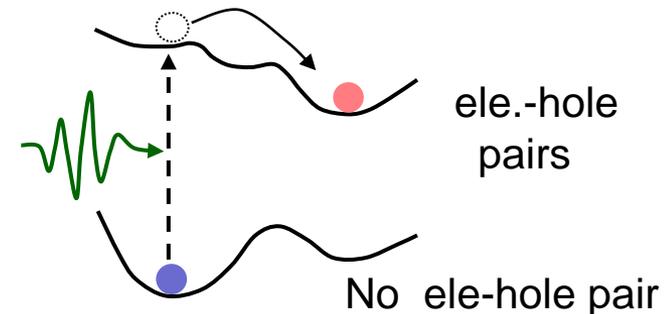


LS/HS mixed state (metal)

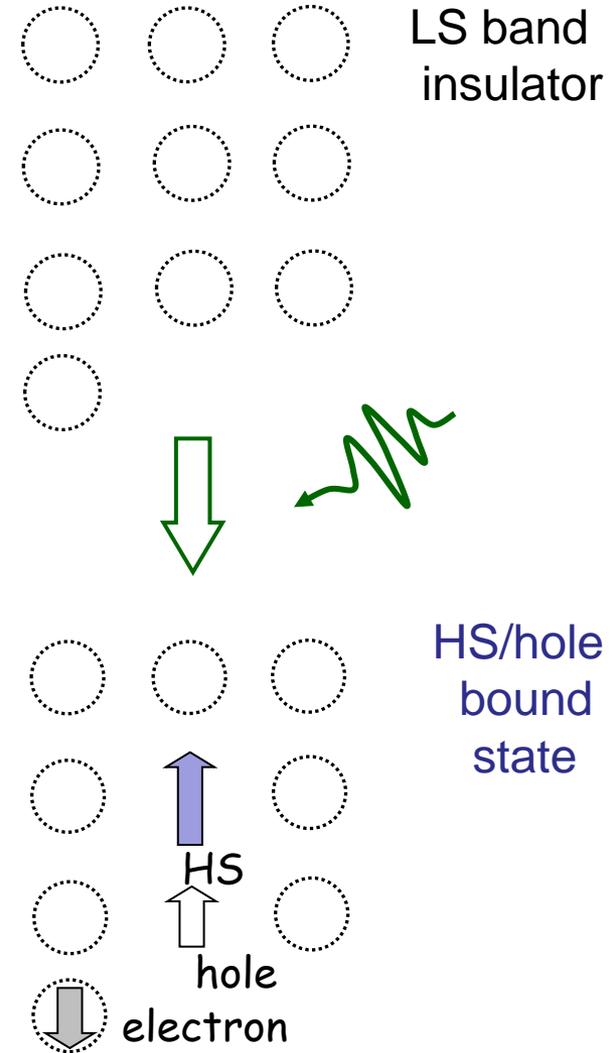
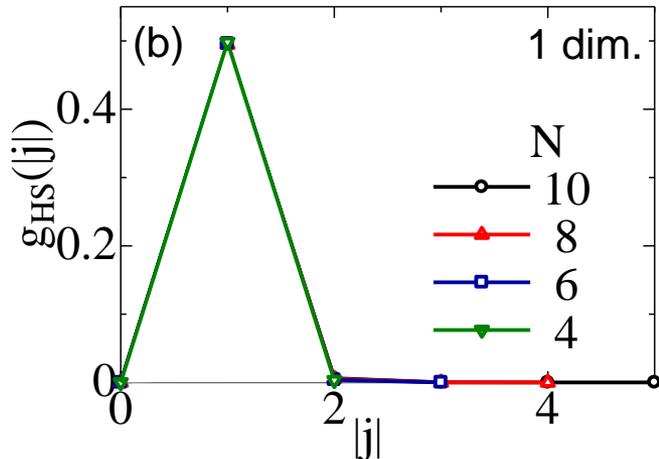
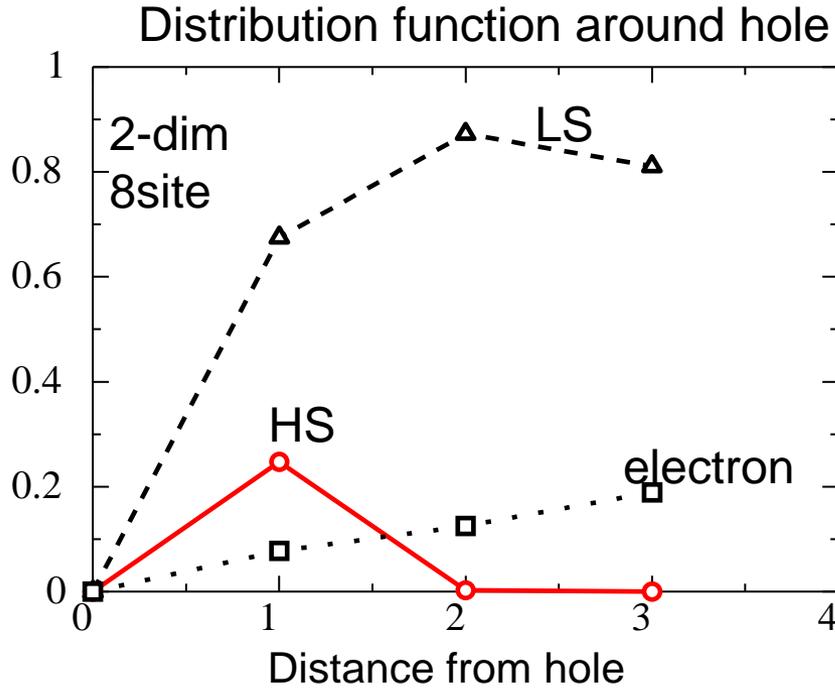
Photoinduced spin state change

■ Lowest energy state

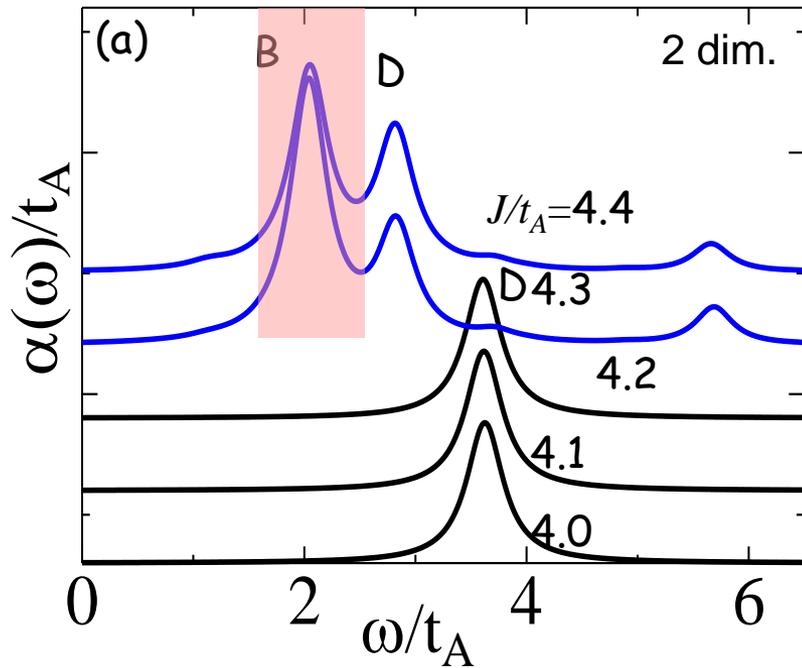
in photo-excited state  
(one ele.-hole pair in N site)



# HS/hole bound state



# Optical pump-probe spectra

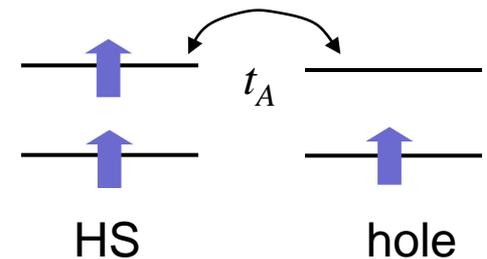
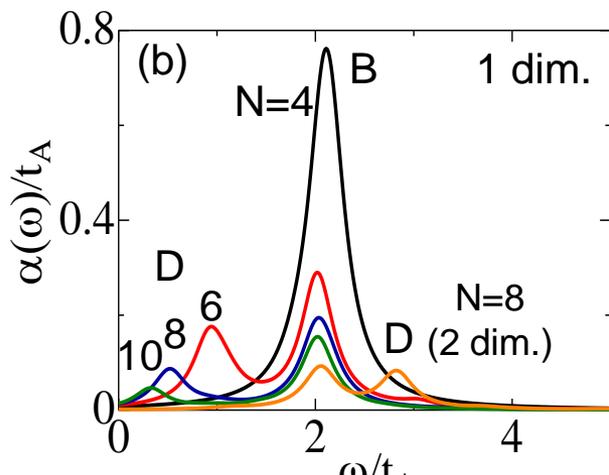
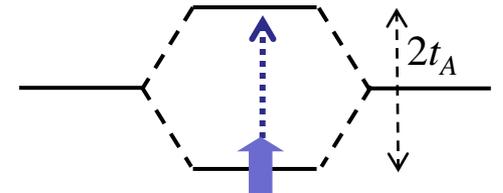


Photoinduced spin-state transition

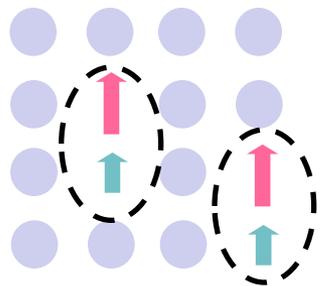
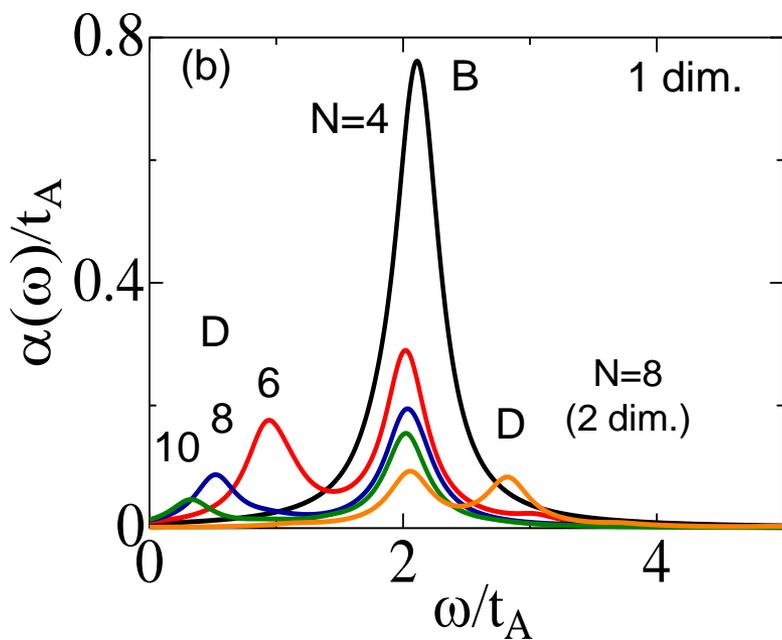
No spin-state transition

- A unique peak structure in photo-induced spin-state transition phase

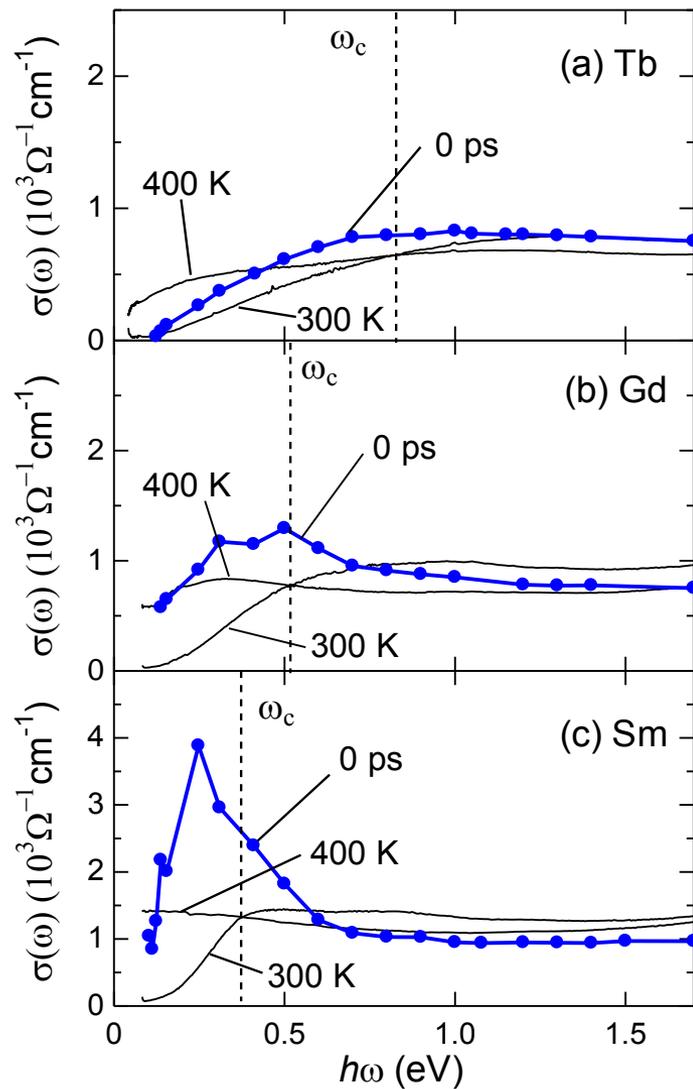
Excitation inside of HS/hole bound state



# Implication for experiments



RBaCO2O6



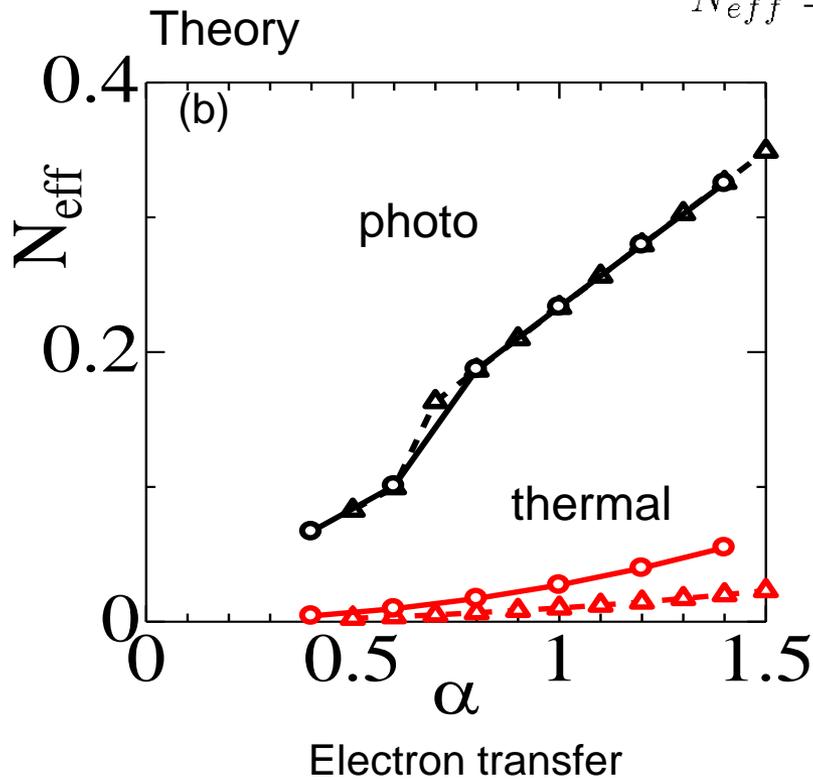
Electron Transfer



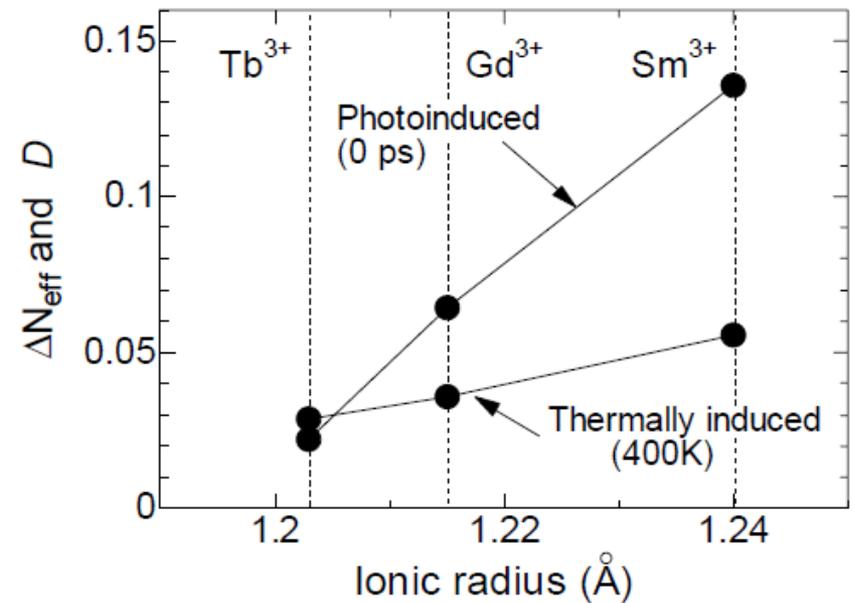
# Implication for experiments

Effective optical carrier number

$$N_{eff} = \int_0^{\omega_c} \sigma(\omega) d\omega$$



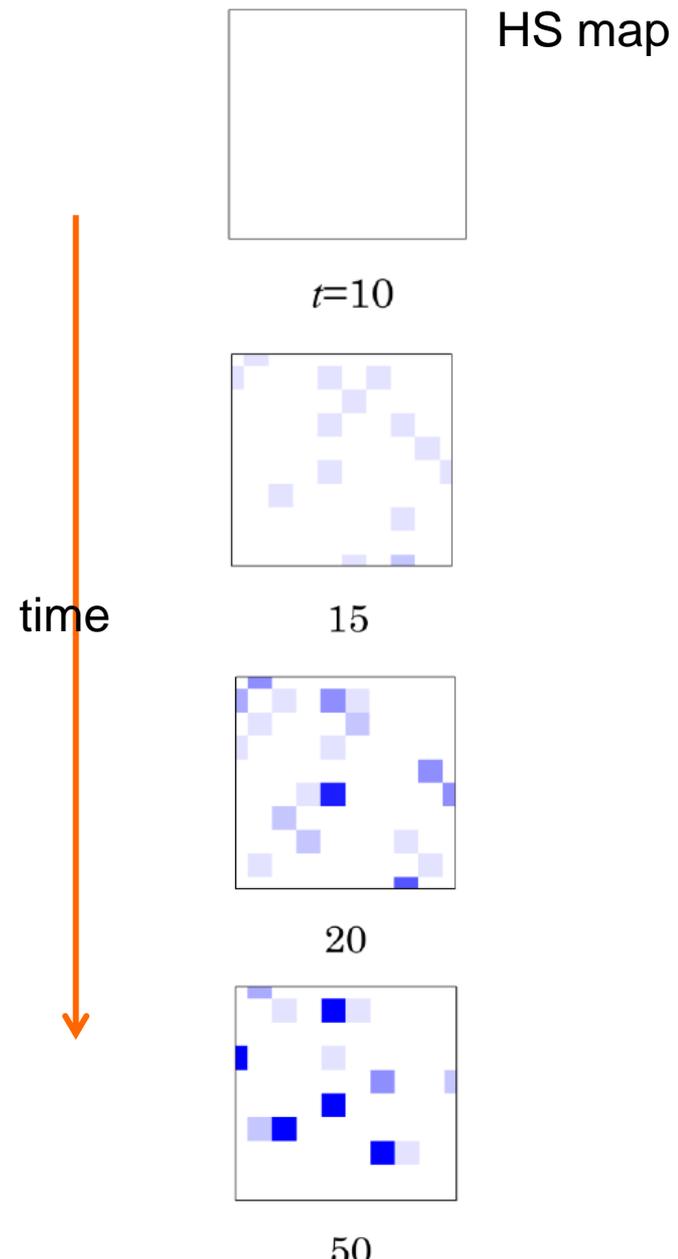
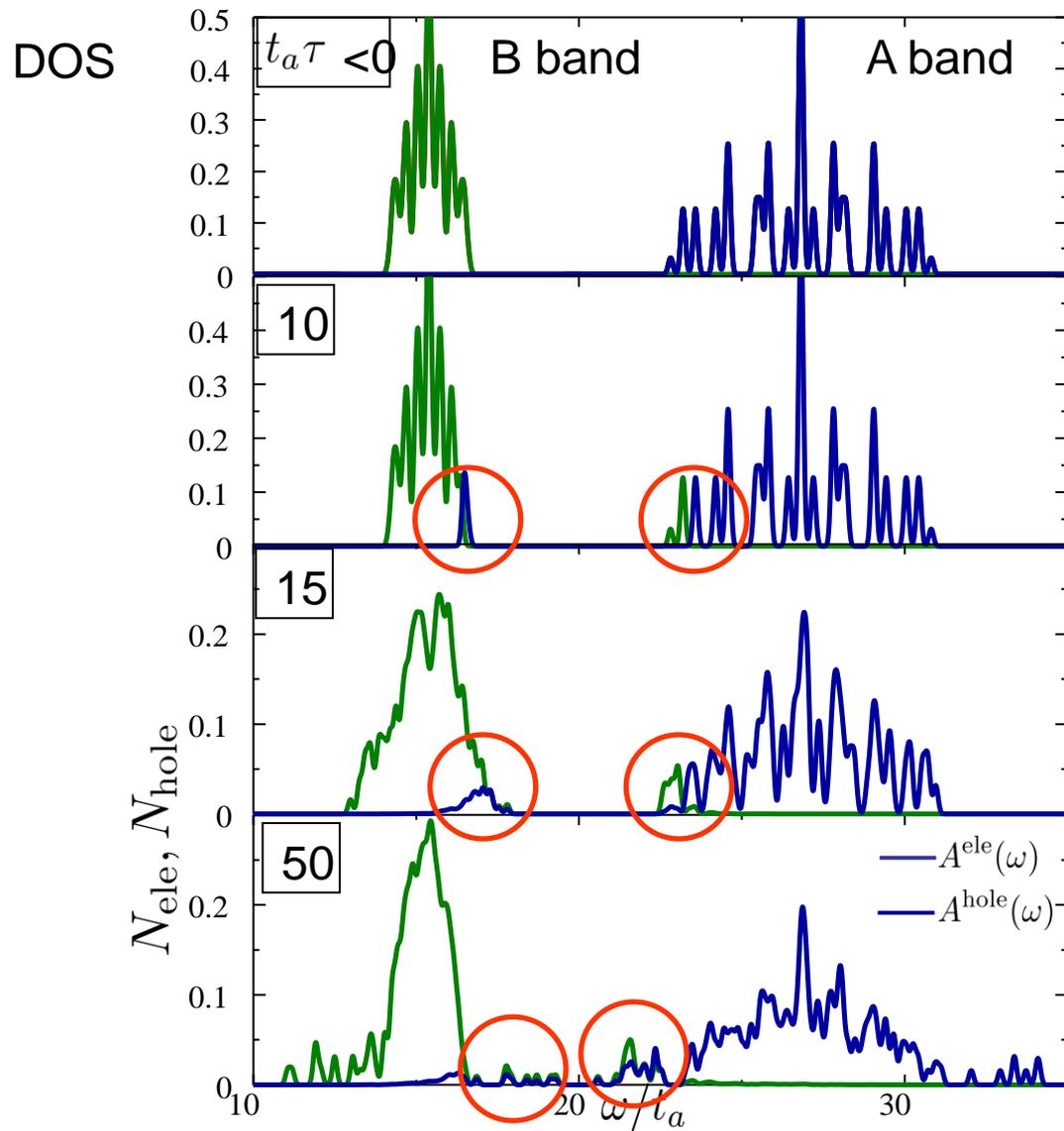
Experiment



Okimoto-Koshihara

$N_{eff}$  increases with transfer  
but  
large difference between thermal & photo

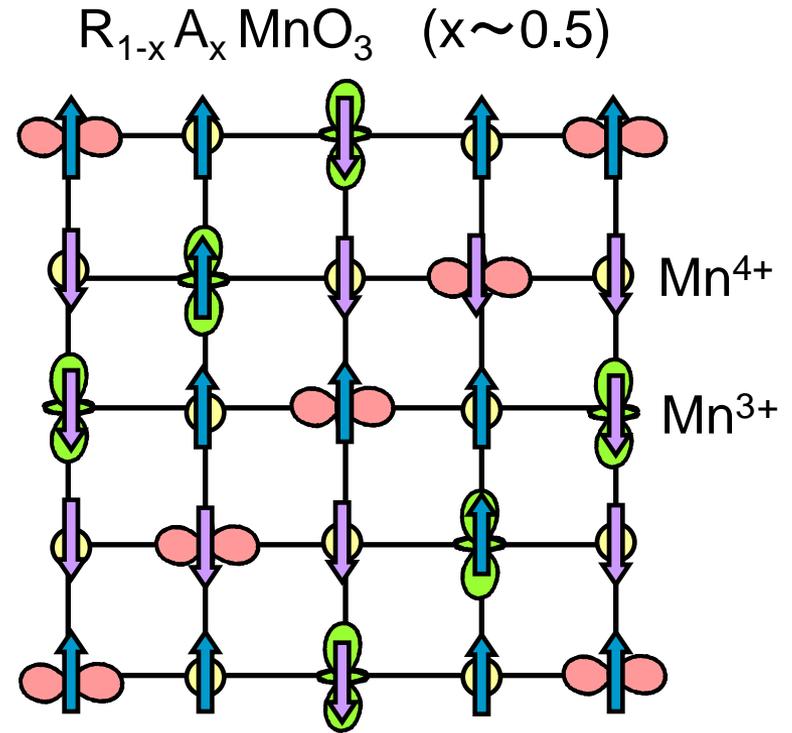
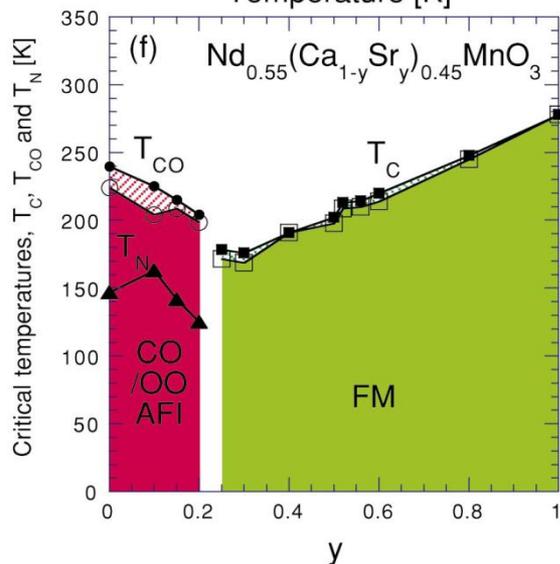
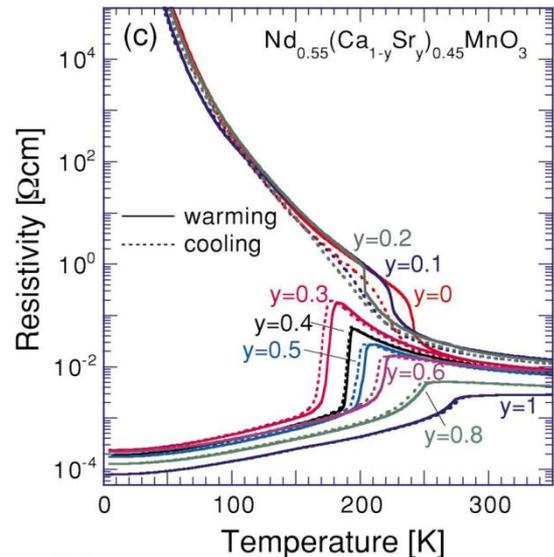
# Time Evolution



# Unconventional photon intensity dependence in Double exchange system

(manganites)

# Manganite: as a charge-spin coupled system



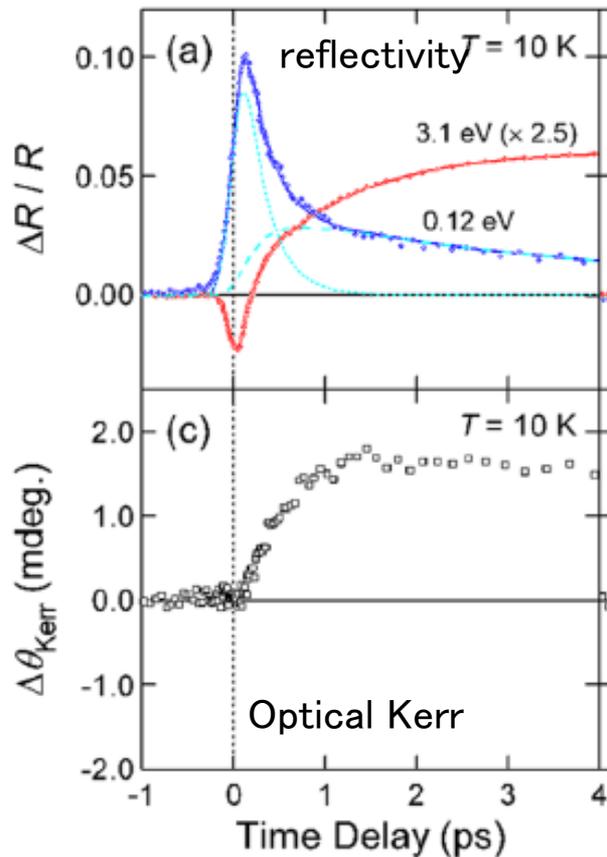
Charge/Orbital/AF  
ordered Insulator  
v.s.  
Ferromagnetic Metal



Colossal Magneto Resistance

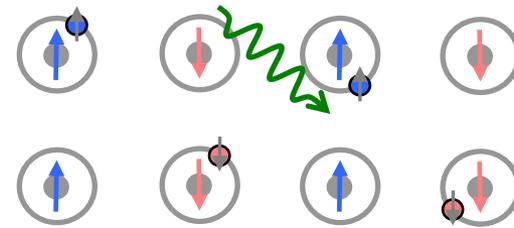
# Photo irradiation

## Optical pump-probe

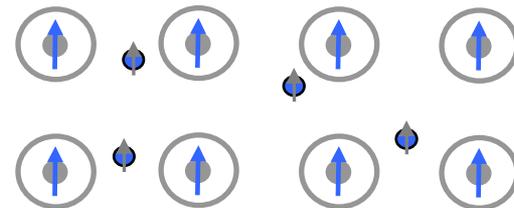


- Photo-irradiation

CO-OO AFM insulator

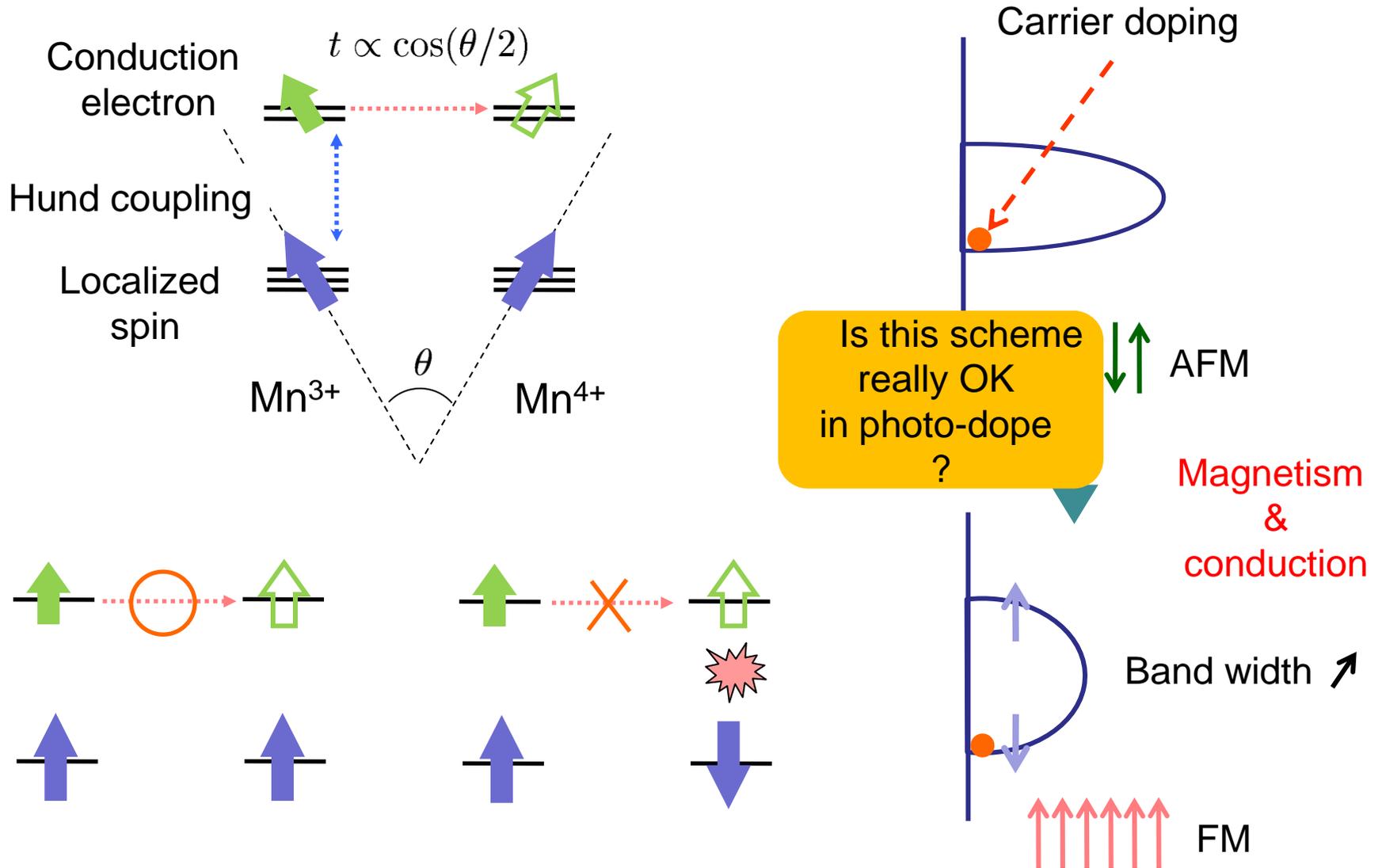


Ferromagnetic metal



# Double exchange interaction

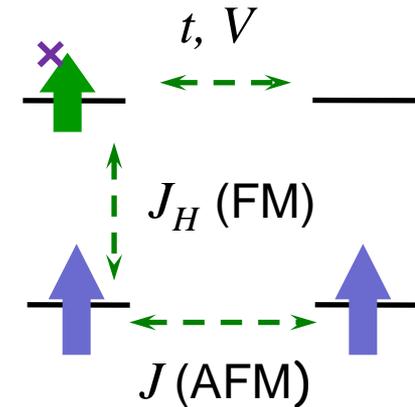
Zener ('51), Anderson-Hasegawa ('55), de Gennes ('59)



# Double exchange model

- double exchange model (ele # / site=0.5)

$$\begin{aligned} \mathcal{H}_{DE} = & t \sum_{\langle ij \rangle \sigma} c_{i\sigma}^\dagger c_{j\sigma} - \frac{J_H}{2} \sum_{iss'} c_{is}^\dagger \vec{\sigma}_{ss'} c_{is'} \cdot \vec{S}_i \\ & + J \sum_{\langle ij \rangle} \vec{S}_i \cdot \vec{S}_j \quad J_H \rightarrow \infty \\ & + V \sum_{\langle ij \rangle} n_i n_j \end{aligned}$$

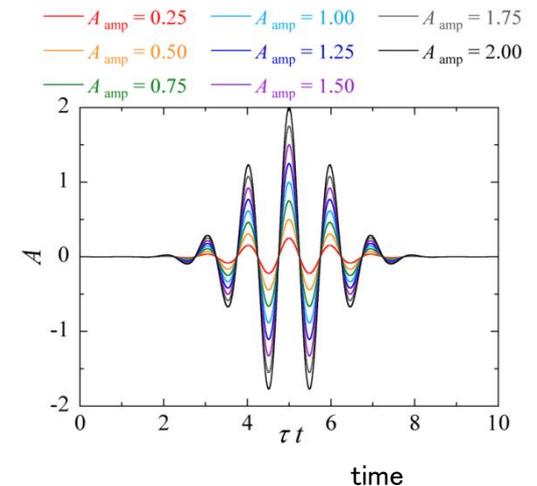


- Pumping

$$t_{ij} \rightarrow t_{ij} e^{-i \int \mathbf{A} \cdot \mathbf{dr}}$$

$$A(\tau) = A_{\text{amp}} e^{-\gamma_0^2 (\tau - \tau_0)^2} \cos \omega (\tau - \tau_0)$$

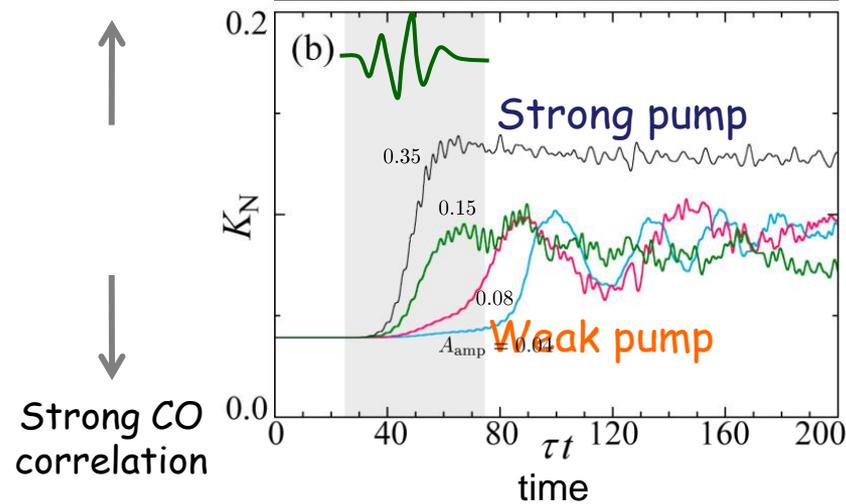
- Exact diagonalization in finite size cluster ( $N \leq 25$ )  
Schrodinger eq. & Bloch eq.



# Charge & spin states

Weak CO correlation  
 Charge correlation func.  $K_N = N^{-1} \sum_{\langle ij \rangle} \langle n_i n_j \rangle$

$10\pi \sim 20F_s$

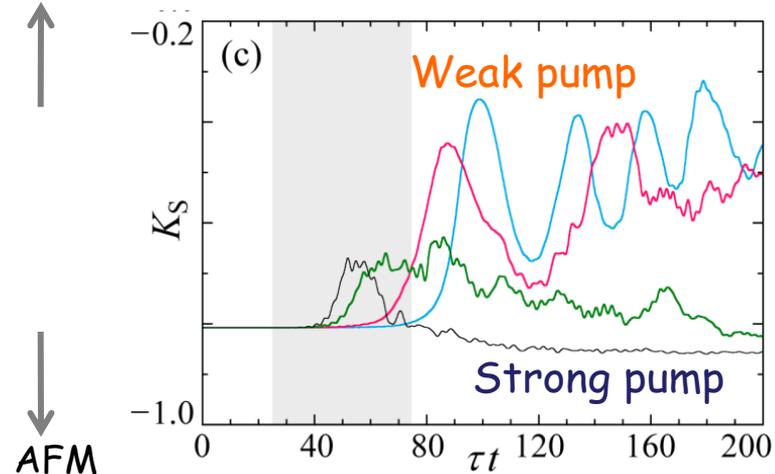


## ■ Melting of CO

Weak pump: Weak melting

Strong pump: Strong melting

FM  
 Spin correlation func  $K_S = N^{-1} S^{-2} \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j$



## ■ Melting of AFM

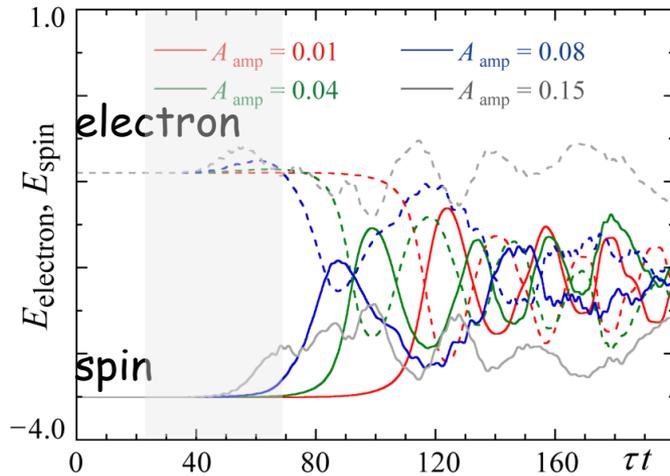
Weak pump: monotonic melting

Strong pump: melting in short time  
 and then  
 increasing of correlation

# Energy flow

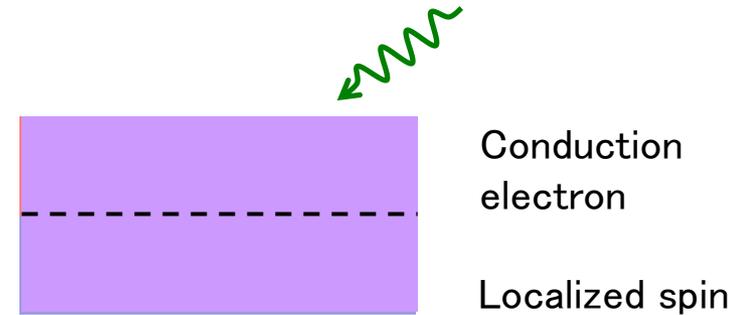
$E_{\text{electron}}$  Kinetic ener. + Coulomb ener..  $E_{\text{spin}}$  Exchange term

## Weak pumping

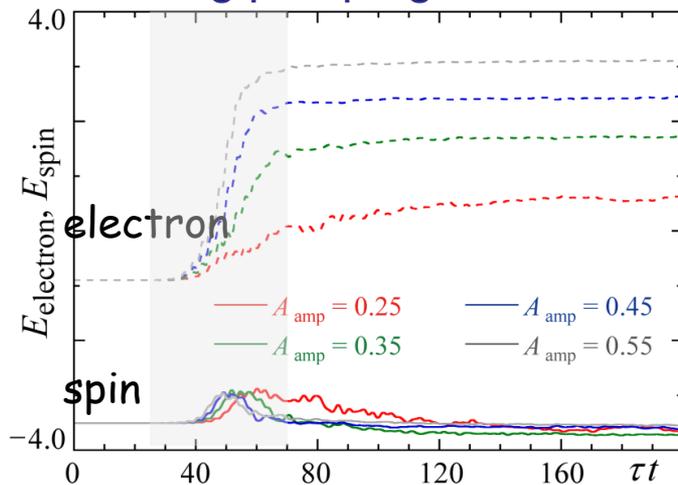


## Weak pump

Electron  $\rightarrow$  Spin



## Strong pumping



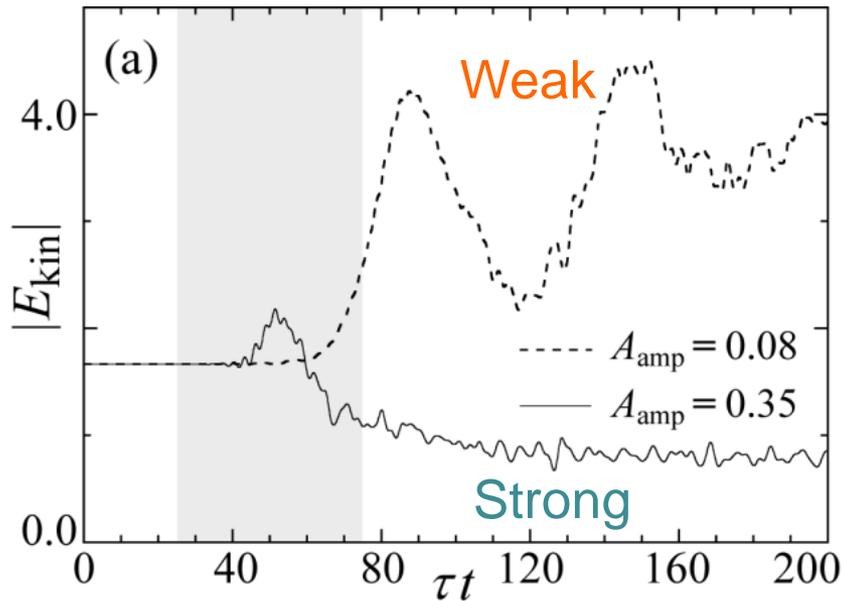
## Strong pump

Spin  $\rightarrow$  Electron

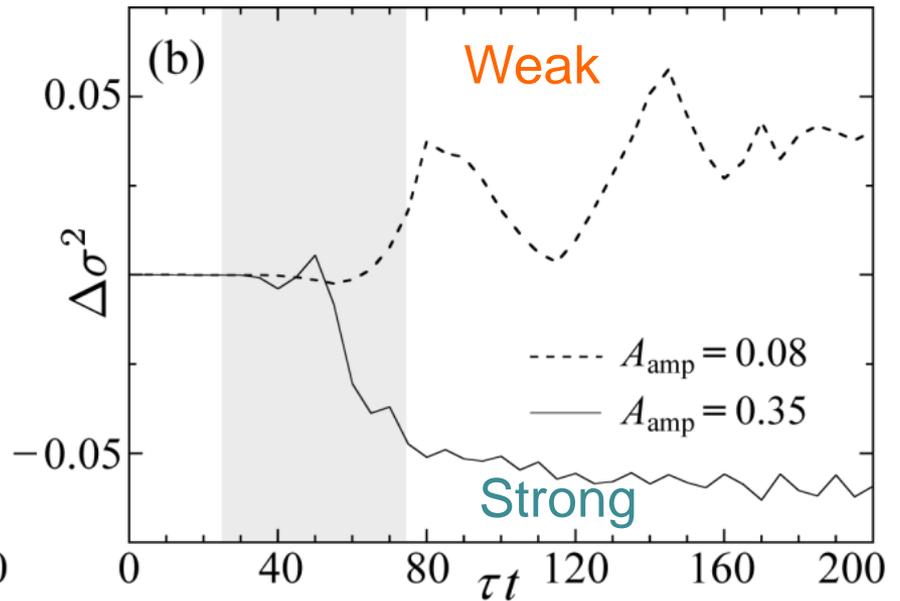


# Strong v.s. weak pumpings

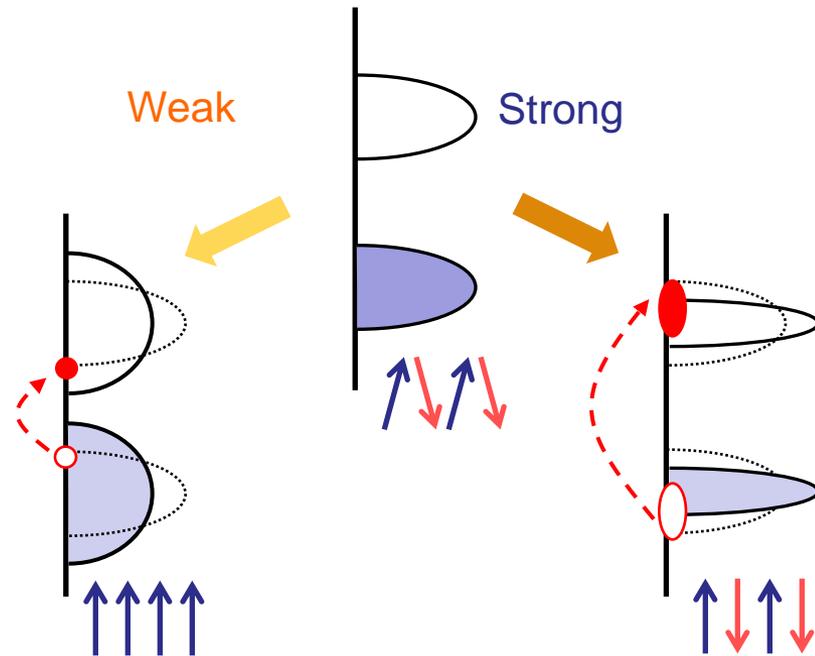
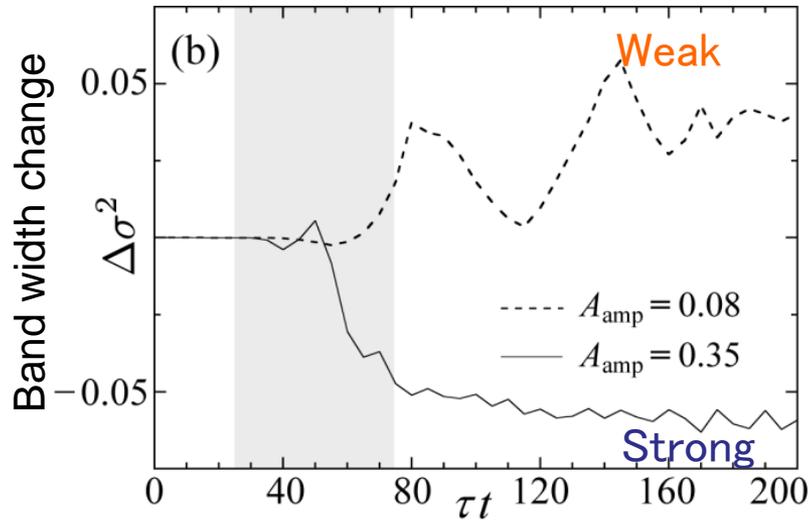
## Kinetic energy



## Bandwidth



# Optical manipulation of band width & magnetism



**Weak** : Increasing of  $W$

Cant CO  $\longrightarrow$  FM metallic  
(conventional double exchange)

**Strong** : Decreasing of  $W$

Cant CO  $\longrightarrow$   
charge disordered AFM insulator  
(Unconventional)

Band width  $\longleftrightarrow$  Spin

High excited state by photon

# Summary

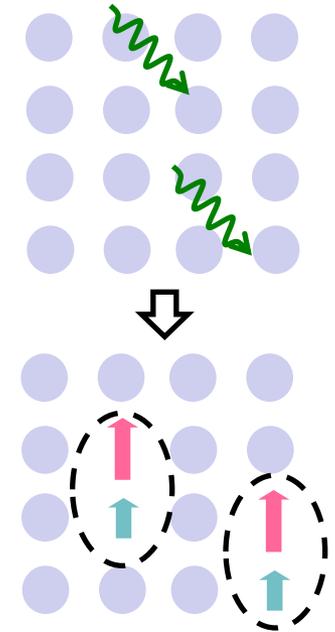
## Optical Manipulation of Magnetism via Double Exchange

### Spin state system (cobaltites)

- Photo-induces spin-state transition
- FM HS-hole bound state
- Much difference from thermal excitation

Y. Kanamori, H. Matsueda and SI,  
Y. Kanamori, J. Ohara and SI

PRL 107, 167403 ('11)  
PRB 86, 045137 ('12)



### Unconventional double exchange (manganites)

- Anomalous photon-density dependence
- **Weak pumping:** CO-AFM  $\rightarrow$  FM metal  
(conventional double exchange scenario)
- **Strong pumping:** Melting of CO but AFM  
(a hidden state)

Y. Kanamori, H. Matsueda and SI  
Y. Kanamori, H. Matsueda and SI,  
J. Ohara Y. Kanamori and SI

PRL 103, 26740 ('09)  
PRB 82, 115101 ('10)  
arXiv.:12041844

